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Ultrafast Chemical Dynamics of Reactions in Beams

6. AUTHOR(S)

Ahmed H. Zewail

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)

California Institute of Technology
1201 E. California Blvd.
Pasadena, California 911258. PERFORMING ORGANIZATION
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13. ABSTRACT (Maximum 200 words)

The research in this proposal focused on the development of femtosecond laser techniques and their applications in the studies of molecular dynamics in real time. The research resulted in some thirty-seven publications with the involvement of more than twenty-five graduate students, post-doctoral fellows, and visiting associates from the U.S. and abroad.

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ULTRAFAST CHEMICAL DYNAMICS OF REACTIONS IN BEAMS

SUMMARY OF WORK ACCOMPLISHED

In the following, we highlight our achievements (and failures) of the work supported by this grant by summarizing the findings presented in some thirty-seven publications of the Caltech group, with the references given in every section.

(A) DEVELOPMENT OF PROBING METHODS

In the previous grant, we outlined the method used for probing the transient behavior of dissociation reactions and reactions restricted to van der Waals complexes (so-called "oriented" bimolecular). In most cases, the probing method depends on the change in the laser-induced fluorescence feature as fragments separate. Since the recoil velocity of fragments is typically 1 km/second, the current state of the art in femtosecond technology (6 fs duration) allows one to view motions, in principle, with a resolution of $\sim 0.06 \text{ \AA}$.

Over the past three years, we have made progress in extending the probing methodology in three directions. First, we built a new molecular beam system that can be housed on a floating optical table with a time-of-flight mass spectrometer (Fig. 1), and this makes it possible to record changes in mass or mass distributions while maintaining the delicate

femtosecond (fs) resolution. Examples for studies by this method involved applications to the dissociation of methyl iodide, iodine,.....etc. by following the evolution of the fragment mass on the fs time scale.

Second, we have extended the range of probing to high energies by using multiphoton excitation/probing, and this made it possible to study the nuclear motion of high energy states as demonstrated by the iodine studies (this group) and the Na clusters (Gerber's group in Freiburg).

Third, for larger molecular systems, we found that the use of depletion techniques is powerful in separating processes such as intramolecular energy redistribution from isomerization or proton transfer. Basically, the intermediate state population is depleted and because of the tunability of the initial laser and the fluorescence of the intermediate state, one is able to follow the changes of the transients, from the initial to the final state.

The new findings of the research done in these areas are discussed in the following publications:

1. Femtosecond Probing of Molecular Dynamics by Mass-Spectrometry in a Molecular Beam.

M. Dantus, M. H. M. Janssen, and A. H. Zewail
Chem. Phys. Lett., 181, 281 (1991)

2. Femtosecond Multiphoton Dynamics of Higher-Energy Potentials.

R. M. Bowman, M. Dantus, and A. H. Zewail
Chem. Phys. Lett., 174, 546 (1990)

3. Femtosecond Real-Time Probing of Reactions. IX. Hydrogen-Atom Transfer.

J. L. Herek, S. Pedersen, L. Bañares, and A. H. Zewail
J. Chem. Phys., 97, 9046 (1992)

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(B) ELEMENTARY REACTIONS

In continuation of our efforts to study the elementary motions in reactions involving two, three, or four atoms, we have focused our attention to studies of the following systems: NaI, NaBr, I₂, Bi₂, and HgI₂. Each of them has unique potential energy feature(s) which allowed us to examine the nature of the dynamics with a particular question in mind. The alkali halides, NaI and NaBr, turned out to be prototype systems for studying the problem of curve crossing and resonance motion as the system changes with time from being covalent to being ionic. We exploited this system to learn how to probe trajectories of the motion as the separation between the two atoms and time increases. It was found that the wave packet motion can be observed going through the probe window in both the positive and negative separation directions, and this made it possible to clock the motion and deduce the potential. Previously, we have been able to determine the coupling matrix element between the covalent and ionic potentials for NaI and NaBr from the decay time of the wave packet. But, surprisingly and accidentally, we found that following the decay of the packet, the coherence of the packet is persistent for up to 40 ps and the resonance motion rephases again. This finding has attracted a number of theoretical studies (V. Engel, H. Metiu, M. Child, and others) and its origin was related to the so-called chaotic motion in a molecular system (see Figs. 2, 3, 4).

For I₂ and Bi₂, the focus was on understanding dissociative and predissociative potential curves (Figs. 5 and 6), while for HgI₂ the focus was on studies of a motion on a potential energy surface (Figs. 7, 8, 9). HgI₂, as a complex, is formed in the bimolecular collision Hg + I₂, as demonstrated by the late Professor R. Bernstein and his group. When

stable HgI_2 is excited, the complex $[\text{HgI}_2]^\ddagger^*$ has at least two vibrational motions that must be considered, the symmetric stretch which leads to the $\text{Hg} + \text{I} + \text{I}$ channel, and the asymmetric stretch which leads to the $\text{HgI} + \text{I}$ product channel. We have examined the femtosecond dynamics of both channels and monitored the behavior as the system descends from the saddle-point region of the transition state to the final products. It was interesting to directly probe the coherent vibrational and rotational motions impacted on the products and to relate these findings to the nature of the potential (bonding, antibonding,.....etc.). From the polarization characteristics of the transients, we were able to deduce the average geometry of the complex $[\text{IHgI}]^\ddagger^*$ (Figs. 7, 8, 9).

These studies of elementary reactions have been extended to different energies and different initial (reactant) and final (products) conditions and the major findings are summarized in the following publications:

1. Femtosecond Real-Time Probing of Reactions. IV. The Reactions of Alkali Halides.
T. S. Rose, M. J. Rosker, and A. H. Zewail
J. Chem. Phys., 91, 7415 (1989)
2. Femtosecond Probing of Persistent Wave Packet Motion in Dissociative Reactions: Up to 40 Picoseconds.
P. Cong, A. Mokhtari, and A. H. Zewail
Chem. Phys. Lett., 172, 109 (1990)
3. Direct Femtosecond Mapping of the Trajectories in a Chemical Reaction.
A. Mokhtari, P. Cong, J. L. Herek, and A. H. Zewail
Nature, 348, 225 (1990)
4. Femtochemistry of the Reaction: $\text{IHgI}^* \rightarrow [\text{IHg}\cdots\text{I}]^\ddagger^* \rightarrow \text{HgI} + \text{I}$.
R. M. Bowman, M. Dantus, and A. H. Zewail
Chem. Phys. Lett., 156, 131 (1989)

5. Femtosecond Real-Time Probing of Reactions. V. The Reaction of I₂HgI.
M. Dantus, R. M. Bowman, M. Gruebele, and A. H. Zewail
J. Chem. Phys., 91, 7437 (1989)
6. Femtosecond Transition-State Spectroscopy of Iodine: From Strongly Bound to Repulsive Surface Dynamics.
R. M. Bowman, M. Dantus, and A. H. Zewail
Chem. Phys. Lett., 161, 297 (1989)
7. Femtosecond Real-Time Probing of Reactions. VI. A Joint Experimental and Theoretical Study of Bi₂ Dissociation.
R. M. Bowman, J. J. Gerdy, G. Roberts, and A. H. Zewail
J. Phys. Chem., 95, 4635 (1991)

(C) WAVE PACKET SPECTRA AND THE POTENTIAL

We have extended the femtosecond studies to bound states. Here, the pulse energy spread is larger than the bound vibrational and rotational energy spacings of the motion. However, if coherent preparation is made, the eigenstates (energies E_n) can be "locked" in phase, according to:

$$\Psi(t) = \sum_n a_n \Psi_n e^{-iE_n t/\hbar},$$

and the observed wave packet motion reflects the involvement of all E_n s in the coherent superposition. By Fourier transforming the temporal data for bound potentials one is able to obtain the high-resolution information on E_n s. Thus, a broad energy pulse is not masking the energy resolution for bound potentials. But, more importantly, the vibrational and rotation motions can be separated in time since the former is on the fs time scale while the latter is on the picosecond or sub-nanosecond time scale. Furthermore, the polarization characteristics of the two motions are

entirely different, one is scalar (vibrational) and the other is vectorial (rotational), and we have used this idea to separate the two and deduce the potential of the motion (Figs. 10, 11, 12).

The following publications summarize our findings:

1. Femtosecond Laser Observations of Molecular Vibration and Rotation.
M. Dantus, R. M. Bowman, and A. H. Zewail
Nature, 343, 737 (1990)
2. Femtosecond Temporal Spectroscopy of ICl: Inversion to the A $^3\pi_1$ State Potential.
M. H. M. Janssen, R. M. Bowman, and A. H. Zewail
Chem. Phys. Lett., 172, 99 (1990)
3. Femtosecond Wave Packet Spectroscopy: Coherences, the Potential, and Structural Determination.
M. Gruebele and A. H. Zewail
J. Chem. Phys. - (January 15, 1993)

(D) THEORETICAL

In order to understand the detailed relationship between the time-resolved data and features of the potential and dynamics, we have invoked classical (and semiclassical) methods, and some quantum calculations using the wave packet approach. With the help of classical trajectories and quantum simulations, we were able to test some of the assumptions regarding the importance of coherence, tunneling,.....etc. For diatoms, the inversion to the potential was based on the RKR method, and for larger systems, we solved the Schrödinger equation in two dimensions and obtained theoretical snapshots of the wave packet to compare with experiments. We have also made some semiclassical approximations to

define bond breaking and bond forming times. The findings are summarized in the following publications:

1. Femtosecond Real-Time Probing of Reactions. III. Inversion to the Potential from Femtosecond Transition-State Spectroscopy Experiments.
R. B. Bernstein and A. H. Zewail
J. Chem. Phys., 90, 829 (1989)
2. Femtosecond Temporal Spectroscopy and Direct Inversion to the Potential: Application to Iodine.
M. Gruebele, G. Roberts, M. Dantus, R. M. Bowman, and A. H. Zewail
Chem. Phys. Lett., 166 459 (1990)
3. From Femtosecond Temporal Spectroscopy to the Potential by a Direct Classical Inversion Method.
R. B. Bernstein and A. H. Zewail
Chem. Phys. Lett., 170, 321 (1990)
4. Femtosecond Real-Time Probing of Reactions. VII. A Quantum and Classical Mechanical Study of the ICN Dissociation Experiment.
G. Roberts and A. H. Zewail
J. Phys. Chem. Special Issue to Professor R. B. Bernstein, 95, 7973 (1991)
5. Femtosecond Wave Packet Spectroscopy: Coherences, the Potential, and Structural Determination.
M. Gruebele and A. H. Zewail
J. Chem. Phys. - (January 15, 1993)
6. Femtosecond Real-Time Probing of Reactions: X. Reaction Times and Model Potentials.
Q. Liu and A. H. Zewail
J. Phys. Chem. Special Issue to Professor D. R. Herschbach - (1993) accepted

(E) COHERENCE, ALIGNMENT, AND CONTROL

One feature of the coherent preparation is the initial control of the phase and the alignment. With polarized pulses, we "induced" a temporal alignment at time zero and followed the change of this alignment with time. This alignment was studied for the IHgI system, the I₂ system, and for the more complex molecular systems discussed below. The results for parallel and perpendicular polarizations help in two ways. First, they give the (coherent) rotational recurrences which provide the rotational constants and the structure; more than 70 structures have been determined by this method. Second, the decay time of the anisotropy can be related to the distribution of angular momenta produced in fragmentation and the qualitative nature of the angular part of the potential. Timing of the sequence of the pulses and their polarization can be used to control the yield of a product channel, as shown for the Xe + I₂ reaction and discussed below. Our findings, so far, have been reported in the following publications:

1. Femtochemistry: The Role of Alignment and Orientation.
A. H. Zewail
J. Chem. Soc., Faraday Trans. 2, 85, 1221 (1989)
2. Femtosecond Real-Time Alignment in Chemical Reactions.
M. Dantus, R. M. Bowman, J. S. Baskin, and A. H. Zewail
Chem. Phys. Lett., 159, 406 (1989)
3. Femtosecond Probing of Persistent Wave Packet Motion in Dissociative Reactions: Up to 40 Picoseconds.
P. Cong, A. Mokhtari, and A. H. Zewail
Chem. Phys. Lett., 172, 109 (1990)

4. Femtosecond Selective Control of Wave Packet Population.
J. J. Gerdy, M. Dantus, R. M. Bowman, and A. H. Zewail
Chem. Phys. Lett., 171, 1 (1990)
5. Femtosecond Laser Control of a Chemical Reaction.
E. D. Potter, J. L. Herek, S. Pedersen, Q. Liu, and A. H. Zewail
Nature, 355, 66 (1992)

(see Figs. 13, 14, 15).

(F) MORE COMPLEX REACTIONS

We have extended the femtosecond reaction dynamics to the domain of large complex systems in an attempt to resolve the primary motions. Of particular interest to us were isomerization (stilbenes) and hydrogen atom transfer (methyl salicylates) reactions. For both, the role of intramolecular vibrational energy redistribution and the subsequent chemical change must be considered.

For systems with a large number of degrees of freedom (N), the situation is more complex than for ABA two-dimensional potentials. First, the one coordinate, perpendicular to the reaction coordinate, is now replaced by the $N-1$ of TS theory. Despite the large N , quantum localization may occur on a dynamic potential. Second, the wave packet motion may suffer fast spreading as its structure is now made of a large number of modes. The isomerization of diphenylethylene (stilbene) is an example of such a complex reaction with 72 modes; the reaction coordinate is described by a single motion about the double bond (torsional angle θ). The molecule at the *cis* configuration is unbound (or quasibound) in the θ coordinate, but, in principle, is bound along all the other coordinates: a saddle point TS in its strict definition.

Recently, we reported the observation of the femtosecond dynamics of the TS in the isomerization reaction of *cis*-stilbene under collisionless conditions. An isolated coherent motion that lasts for several hundred femtoseconds and depends on the total excess vibrational energy has been observed. Such motion correlates with the topology of the potential energy surface (PES) and represents a *nonstatistical* behavior. The experimental results were compared with calculations on a model PES for the reaction. Similar studies were made on methyl salicylate where the motion of the hydrogen atom was found to take place in 60 fs. After tautomerization, the new form undergoes hydrogen bond dissociation with an energy threshold determined by energy redistribution (Figs. 16, 17, 18, 19). The following publications summarize our initial efforts in this new direction:

1. Femtosecond Real-Time Probing of Reactions. IX. Hydrogen-Atom Transfer.
J. L. Herek, S. Pedersen, L. Bañares, and A. H. Zewail
J. Chem. Phys., 97, 9046 (1992)
2. Femtosecond Vibrational Transition-State Dynamics in a Chemical Reaction.
S. Pedersen, L. Bañares, and A. H. Zewail
J. Chem. Phys., 97, 8801 (1992)

(G) GENERAL REPORTS

In several review articles from this group, or in collaboration with other colleagues, the above results and their general implications have been discussed (previous reviews related to this work were given in the last renewal grant):

1. Ultrafast Molecular Reaction Dynamics in Real-Time: Progress Over a Decade.
L. R. Khundkar and A. H. Zewail
Ann. Rev. Phys. Chem., 41, 15 (1990)
2. Ultrafast Reaction Dynamics.
M. Gruebele and A. H. Zewail
Phys. Today, 43, 24 (1990); Ber. Bunsenges. Phys. Chem. (German), 94, 1210 (1990); Parity (Japanese), 5, No. 12, 8 (1990); Sov. Phys. Uspekhi (Russian), 161, No. 3, 69 (1991)
3. Lasers and Ultrafast Phenomena.
A. H. Zewail
in: Lasers in Science and Technology (chapter) - submitted for publication
4. The Birth of Molecules.
A. H. Zewail
Scientific American, 263, 76 (1990); translated to 12 languages.
5. Femtosecond Transition-State Dynamics.
A. H. Zewail
in: *Structure and Dynamics of Reactive Transition States*
Faraday Discuss. Chem. Soc., 91, 207 (1991)
6. Femtochemistry: Recent Advances and Extension to High-Pressures.
A. H. Zewail, M. Dantus, R. M. Bowman, and A. Mokhtari
J. Photochem. Photobiol. A: Chem., 62/3, 301 (1992) - Invited Article

TOF-MS and LIF Molecular Beam

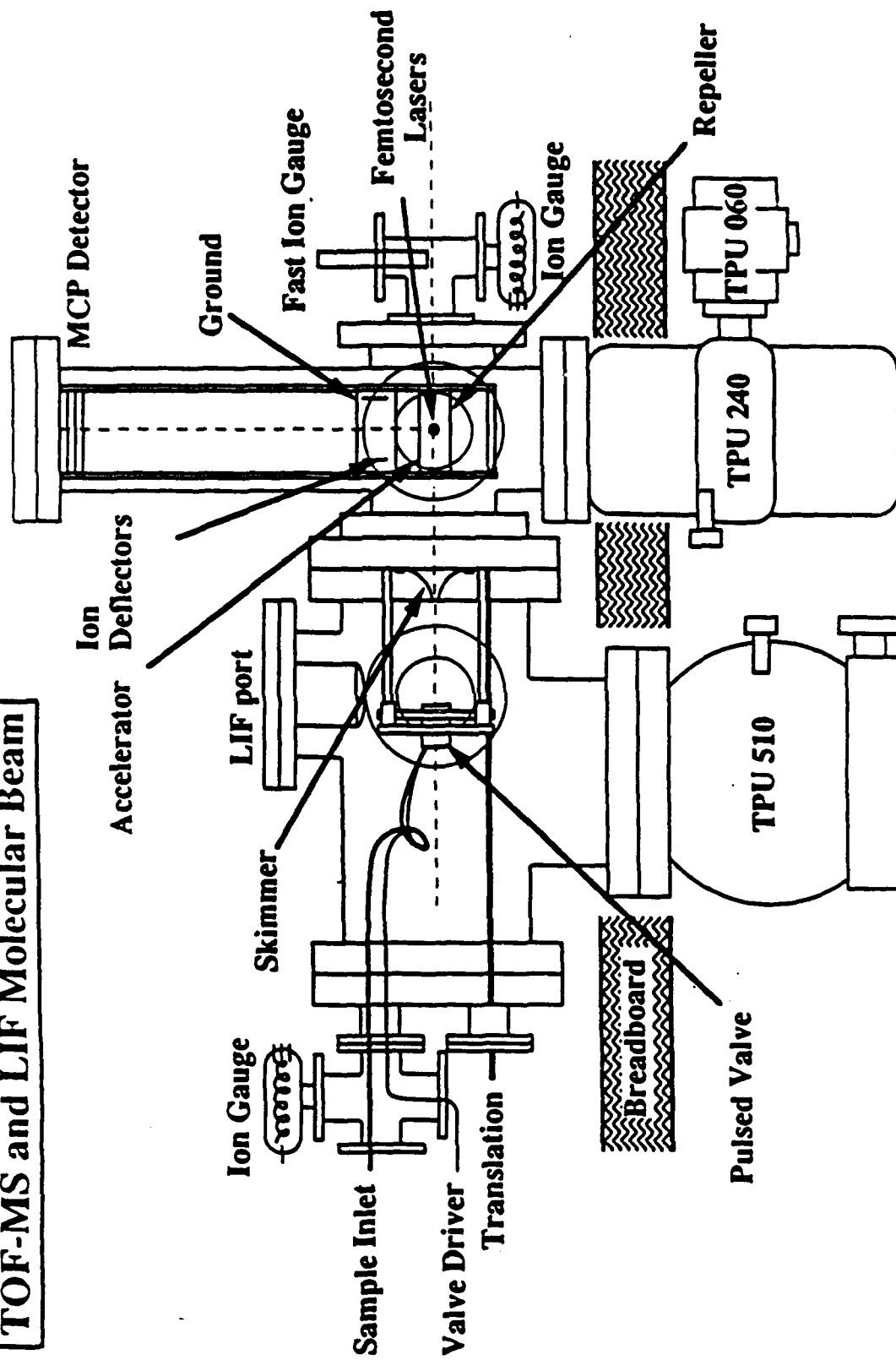


Fig 1 : Molecular Beam apparatus, with TOF and LIF detections, and FS lasers.

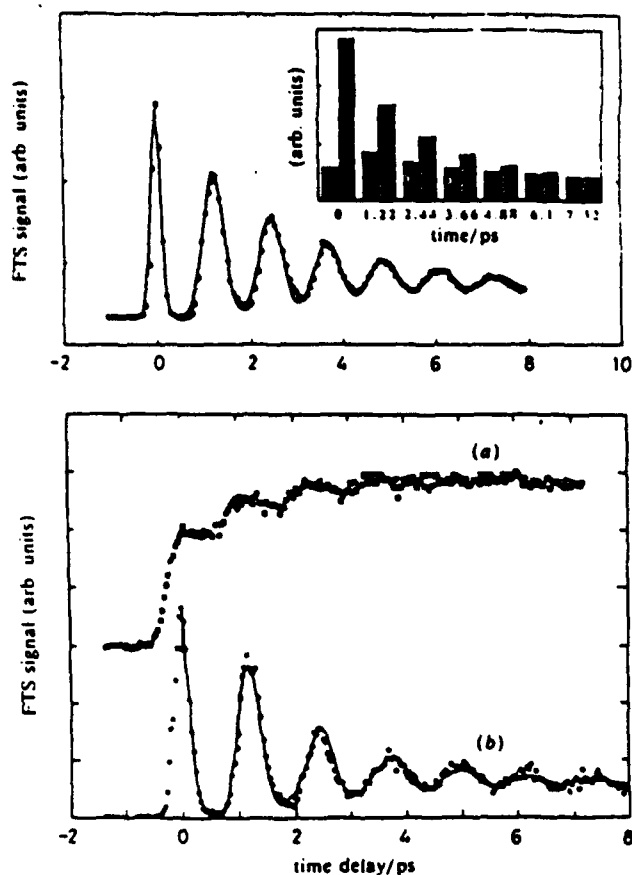


Fig 2: The oscillatory behavior observed by detecting $[Na \cdots I]^{\ddagger\ddagger}$ or Na. The insert shows the delay of the peak height and area of oscillation

Wavepacket motion

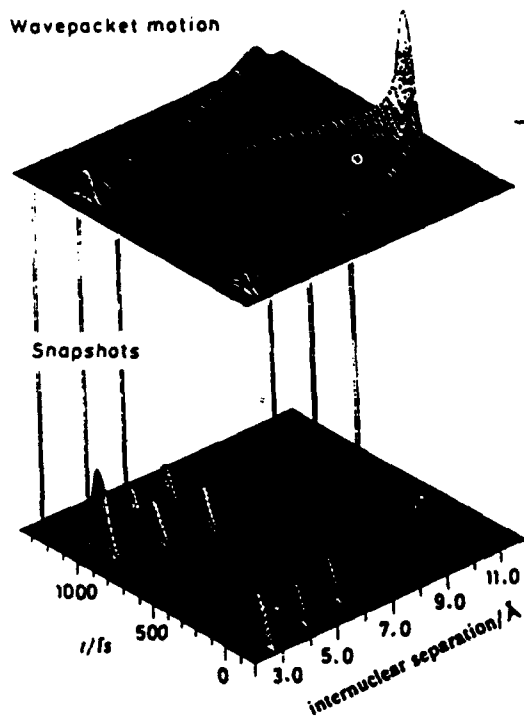


Fig 4a: Theoretical simulation of the trajectory and predicted splitting of resonance peaks.

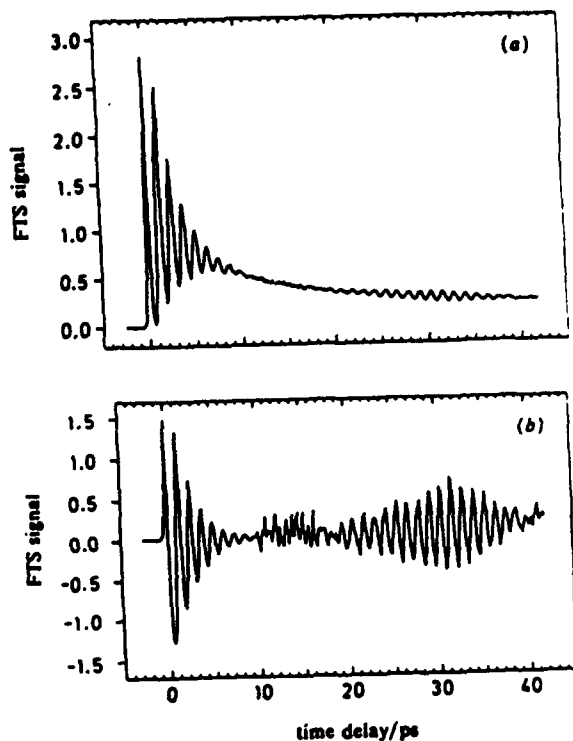


Fig 3: The oscillatory behavior at long times (upto 40 ps) and the rephasing (echo) due to wave packet coherence.

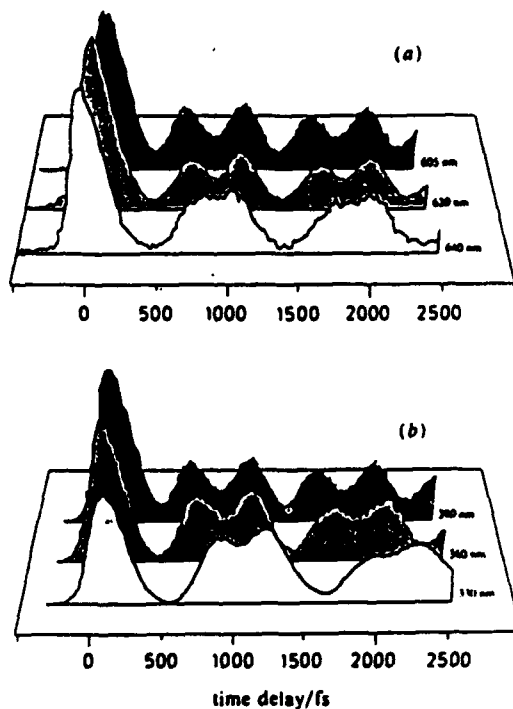


Fig 4b: Experimental observation of the splitting.

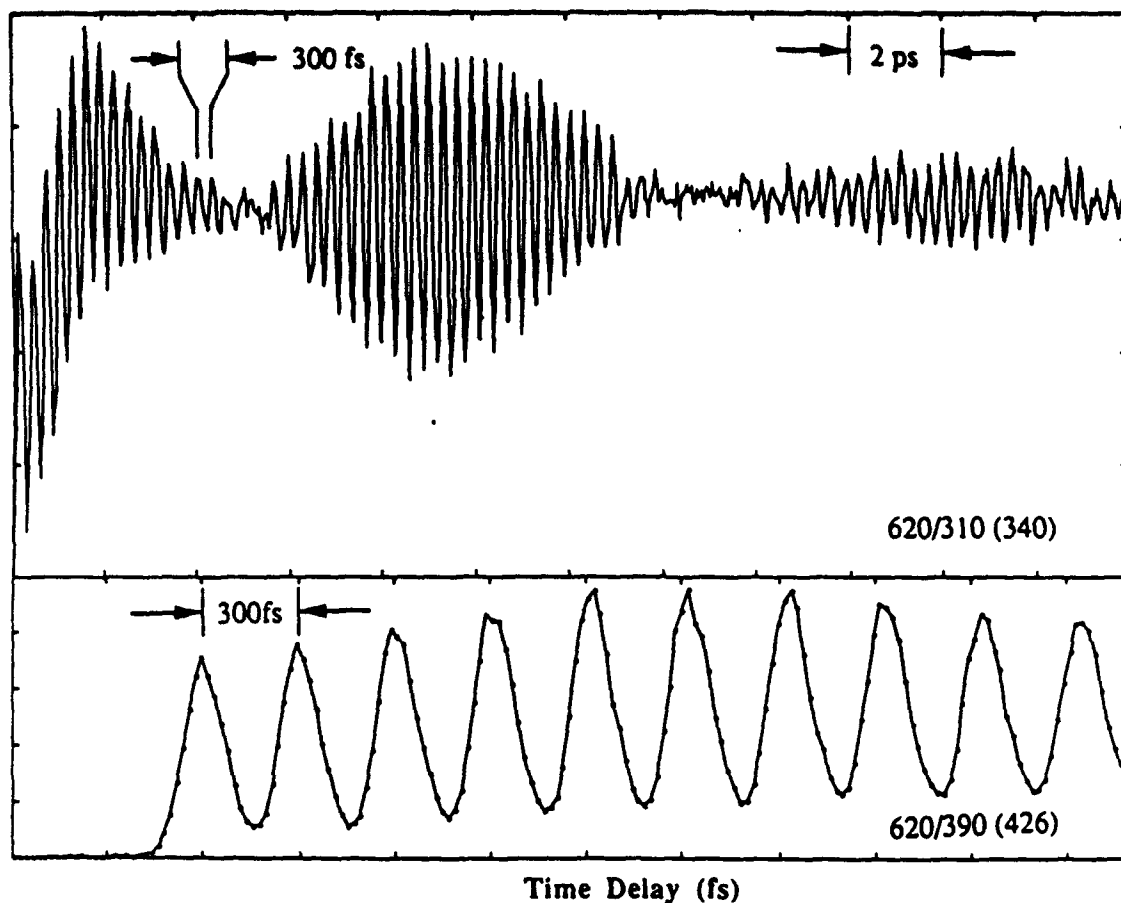


Fig 5a: Typical femtosecond transients for a bound potential (I_2 , B-state) with frequencies indicating the anharmonic nature of the potential.

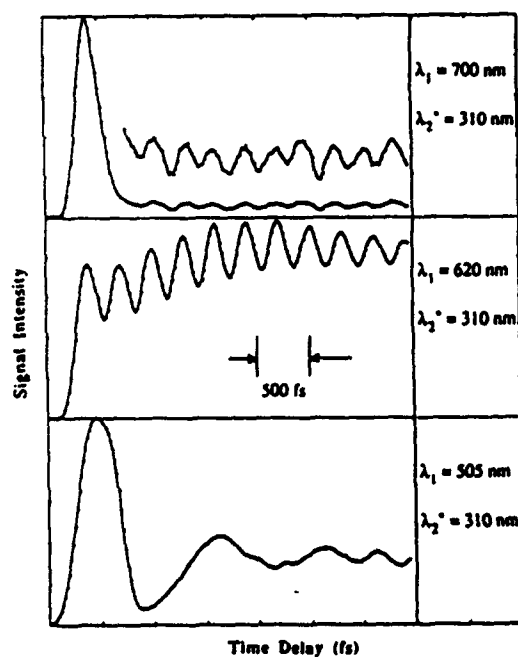


Fig 5b: The change in the transients as the excitation energy reaches up or below dissociation

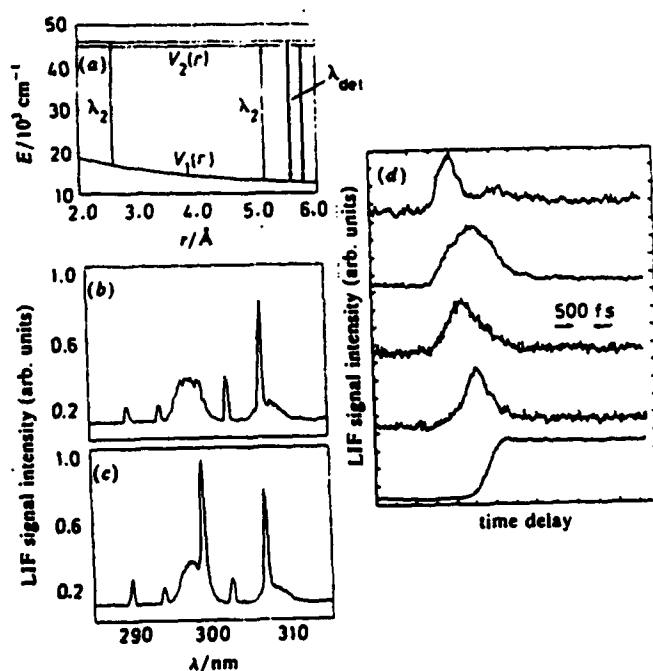


Fig 6: The femtosecond transients of the Bi_2 system, showing transient and final Bi charge with time.

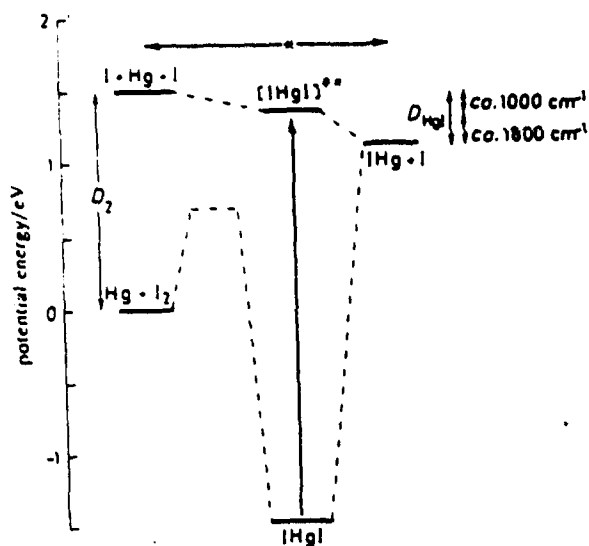


Fig 7a: The energetics for IHgI and $\text{Hg} + \text{I}_2$.

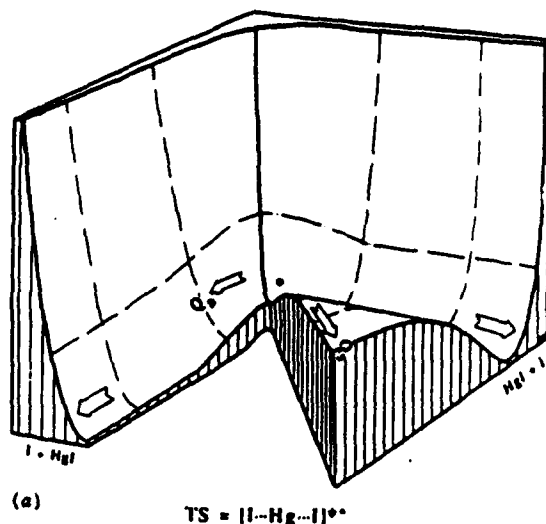


Fig 7b: The symmetric and antisymmetric coordinates of the potential.

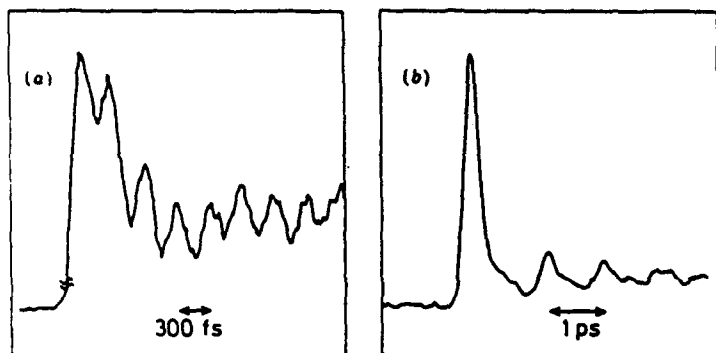


Fig 8a: The scalar transients of IHgI taken for two different final product state.

Fig 8b: The vectorial (polarization) for \parallel and \perp detections.

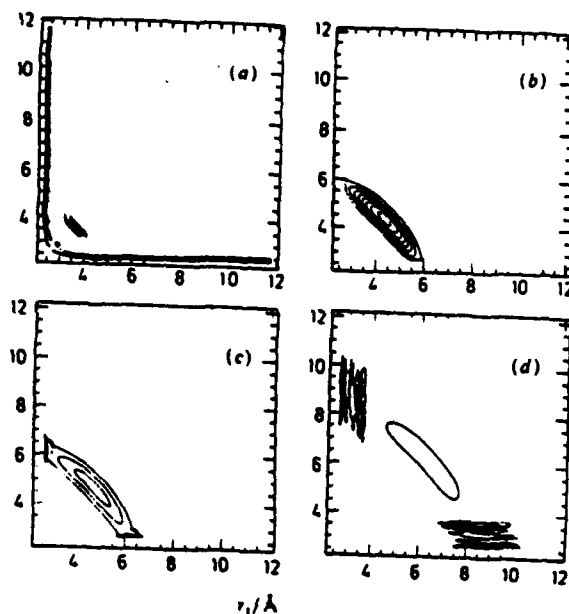
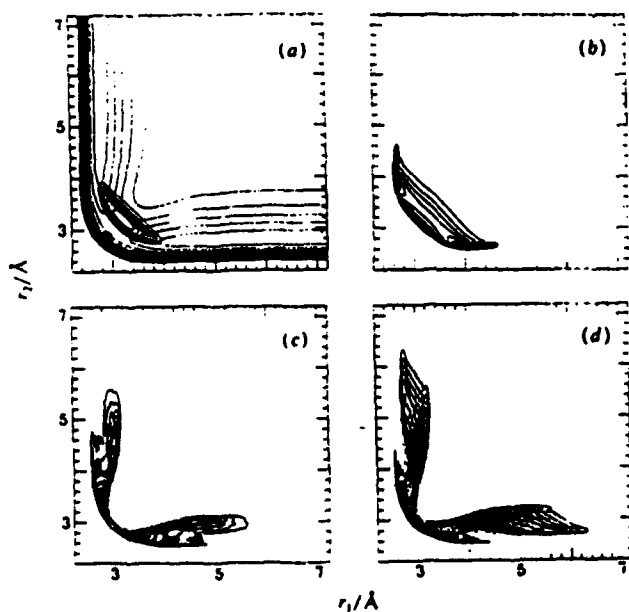
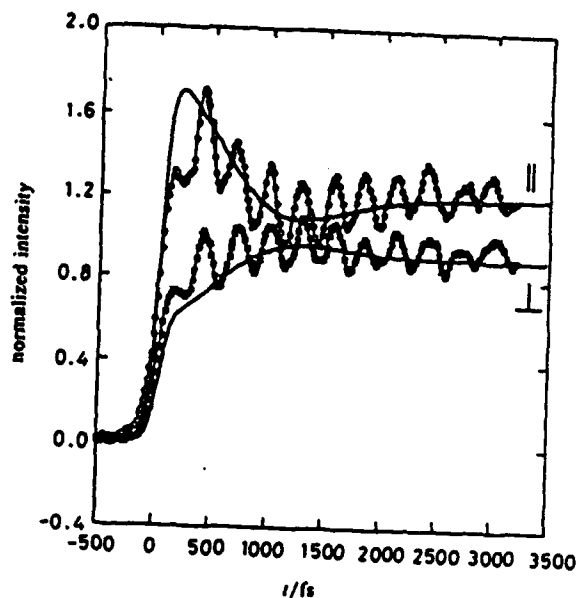


Fig 9: Theoretical snapshots of the wave packet, at two energies, with time.

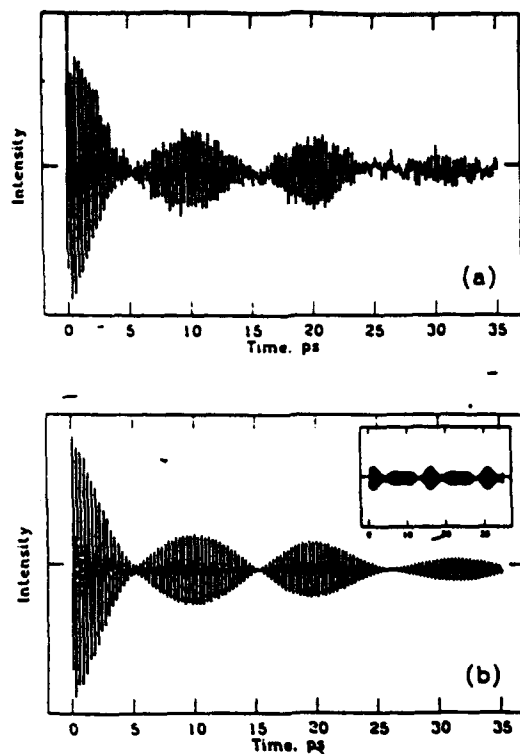


Fig 10 : vibrational wave packet of I_2 (experimental, top; theory bottom)

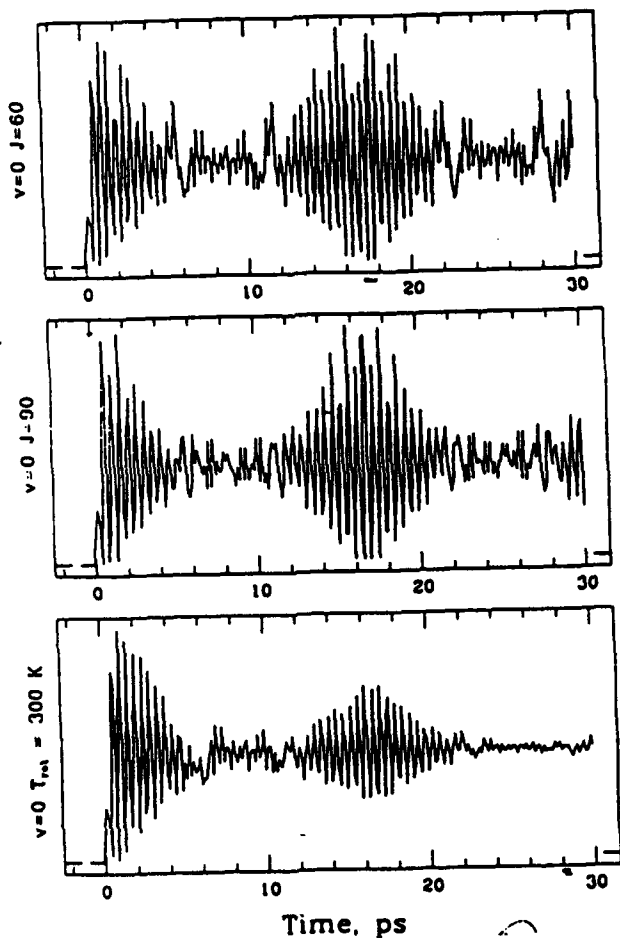


Fig 12a : Quantum simulation of the motion for different initial J .

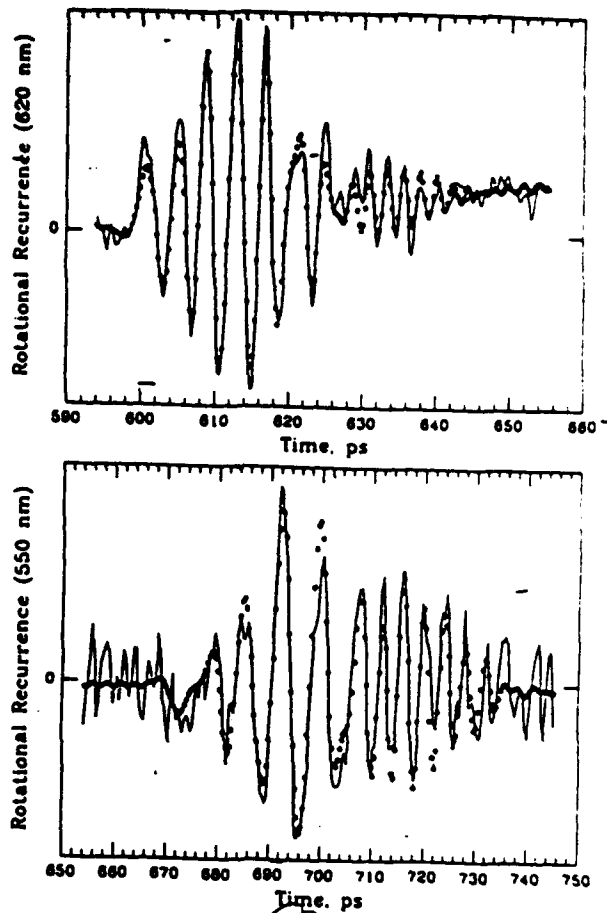


Fig 11 : Rotational wave packet recurrence at two different λ s.

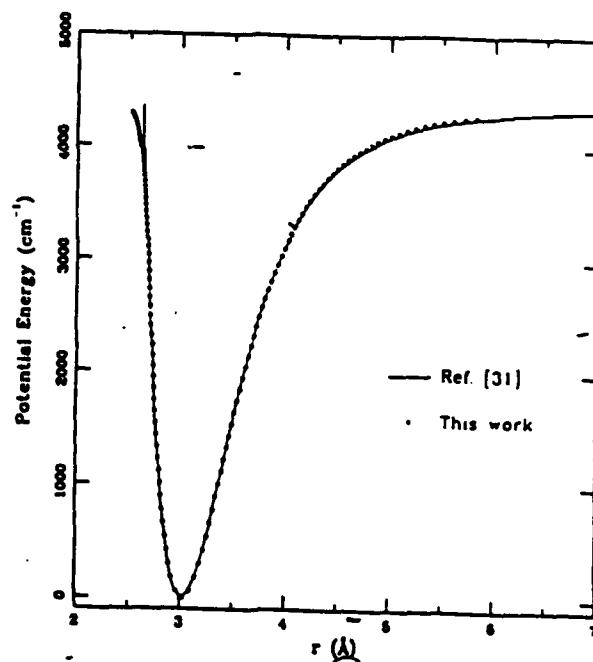


Fig 12b : The (inverted) potential obtained from the experimental results.

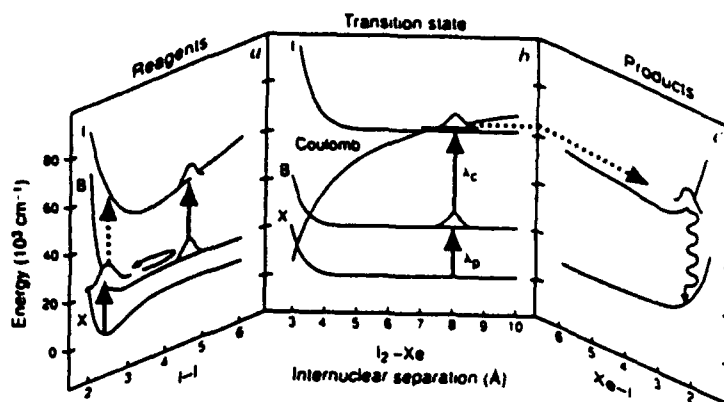


Fig 13 : cuts of the PES along the relevant coordinates for $\text{Xe} + \text{I}_2$ reaction.

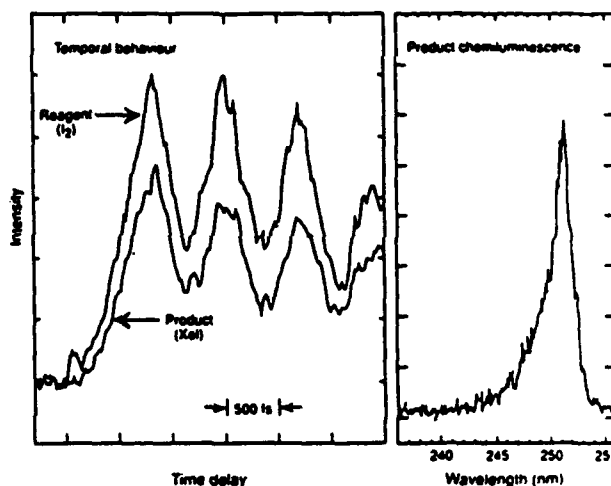
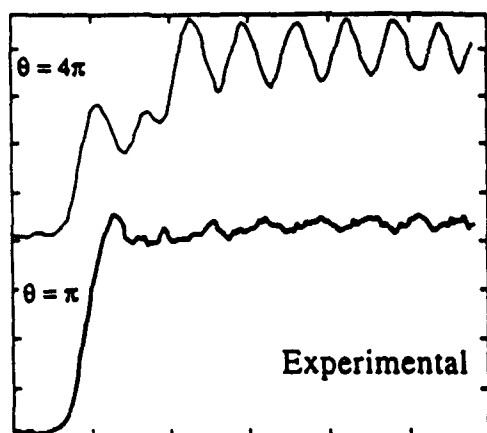


Fig 14 : Experimental results of the yield vs. time.

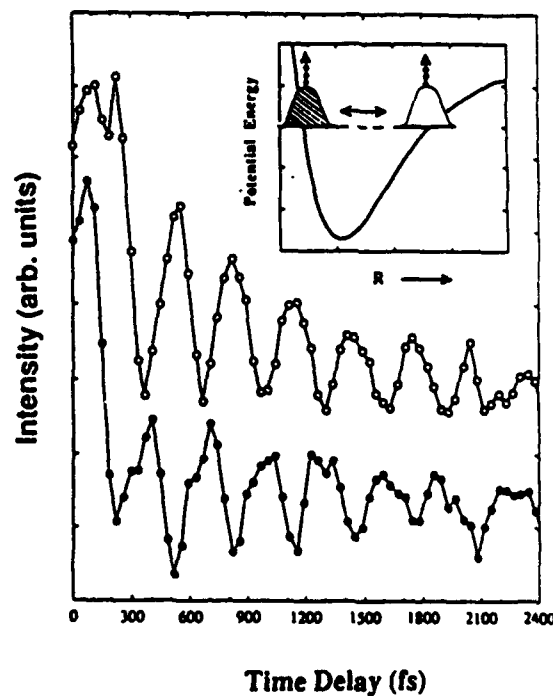
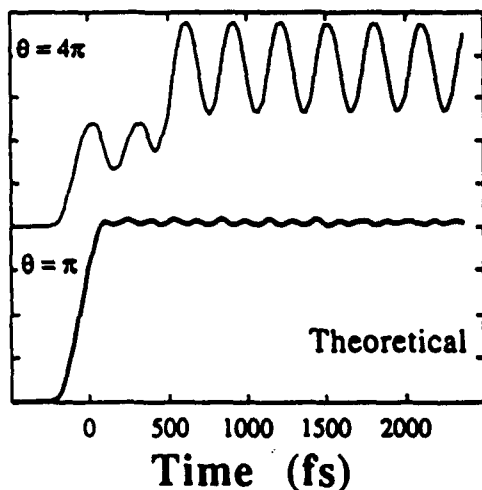


Fig 15a: change in wave packet population as a function of θ , the angle determined by the delay time between the first two pulses.

Fig 15b: The change in phase of the wave packet.

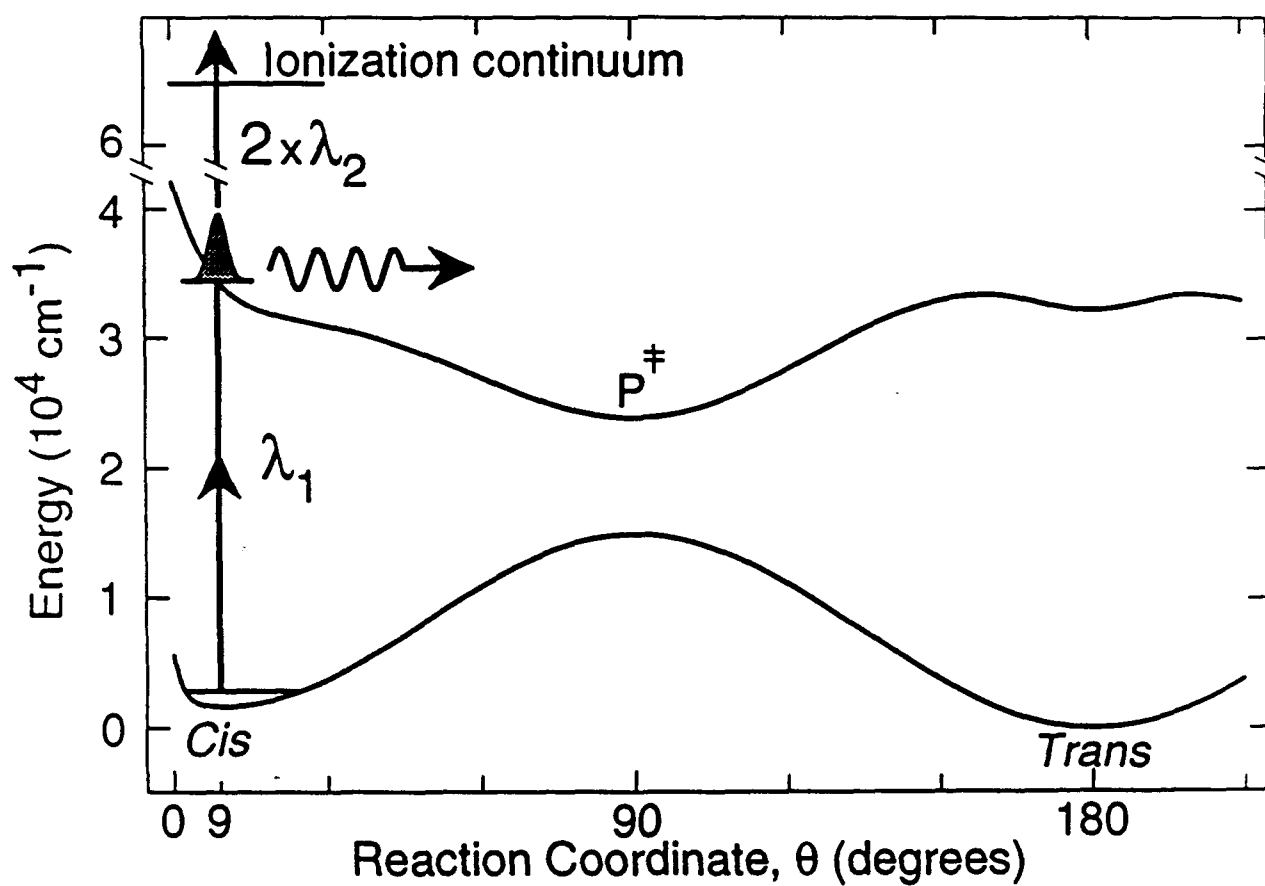
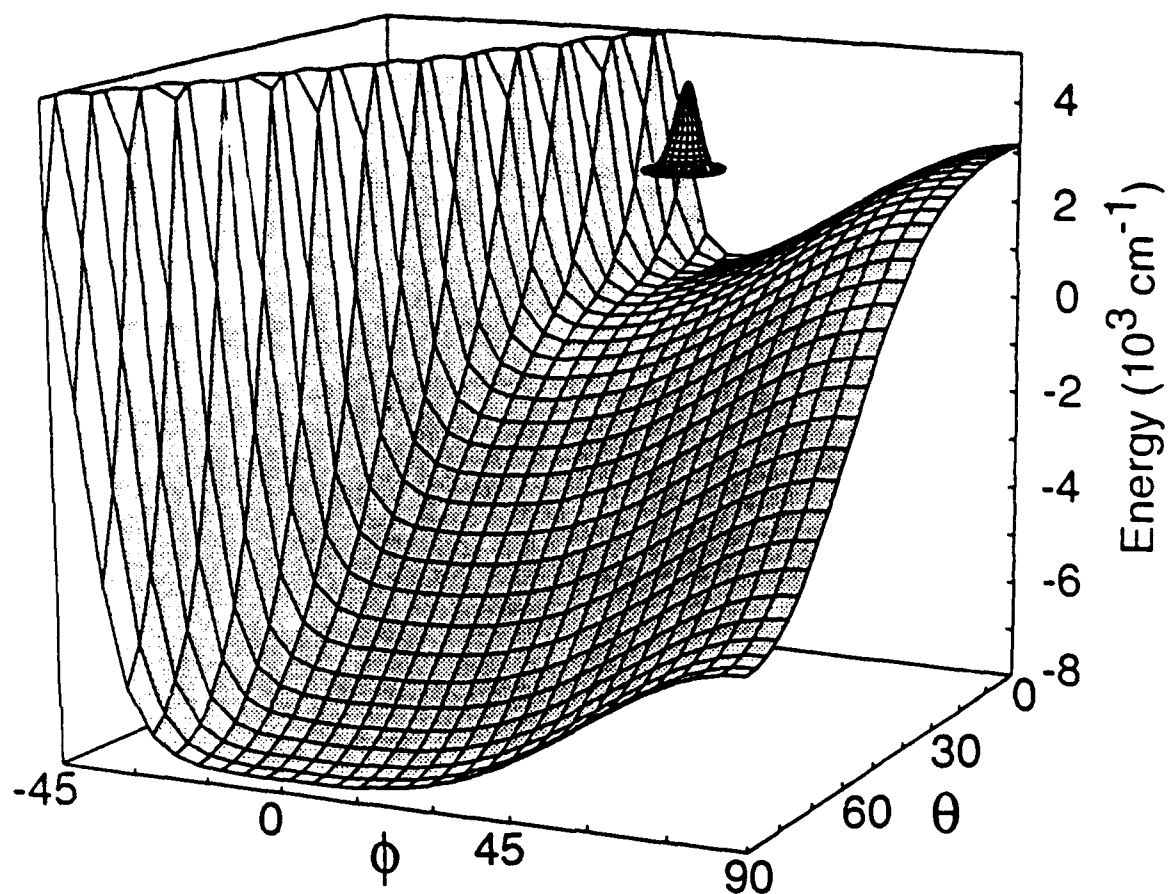


Fig 16: The isomerization potential of stilbene, *cis* and *trans*.

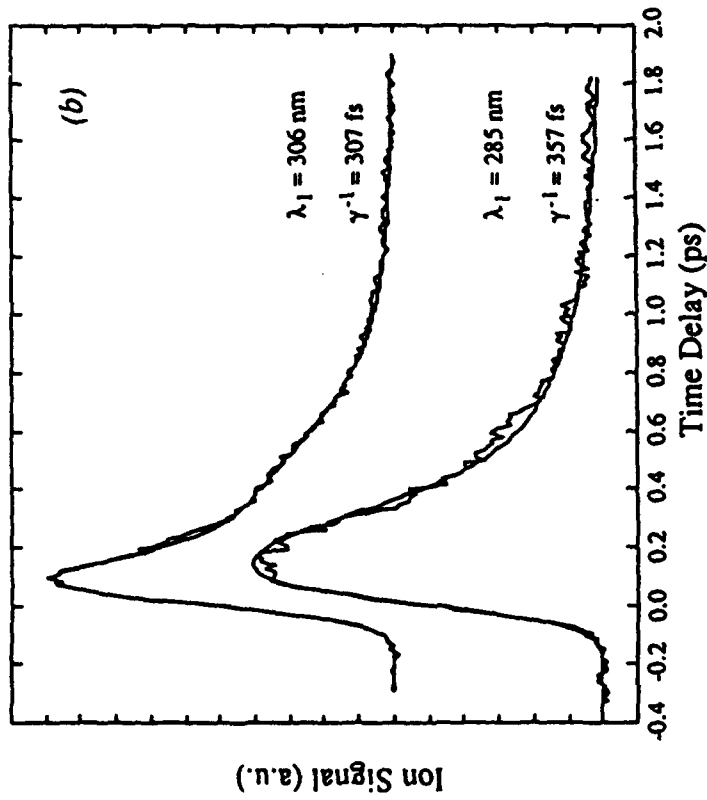
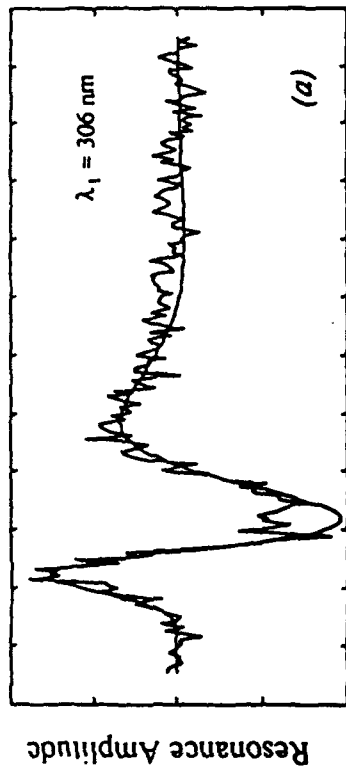


Fig 17a: Experimental fs transients of cis-stilbene.

Fig 17b: The potential at different twisting angles.

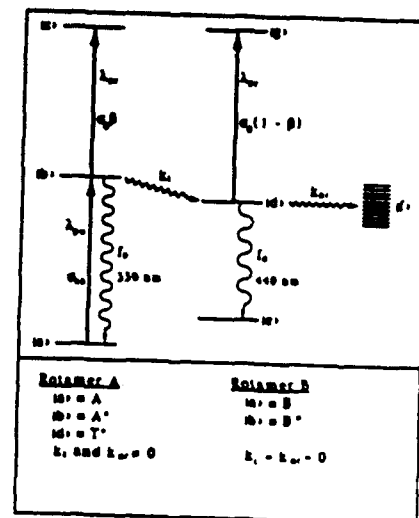
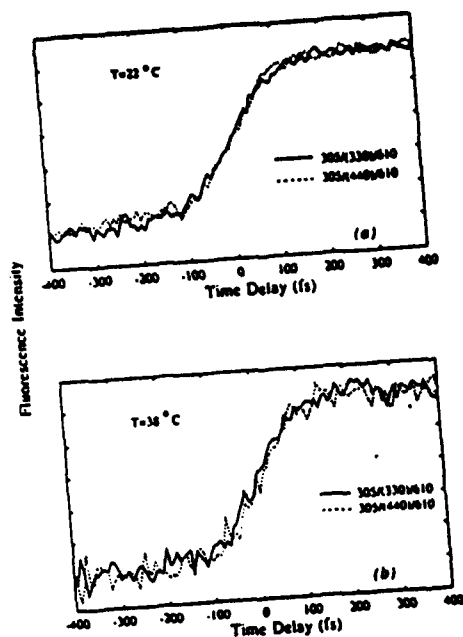


Fig 18 a: fs transients of H atom transfer Fig 18 b: Experimental scheme

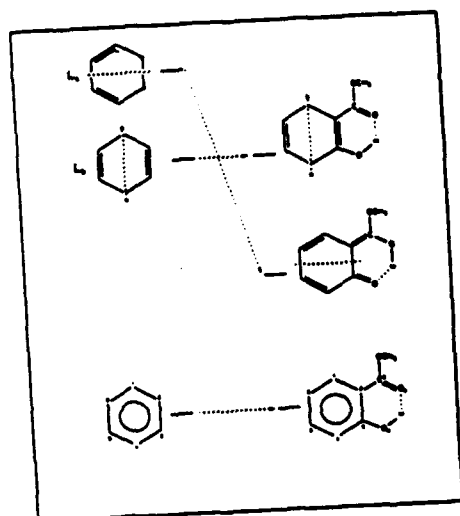


Fig 19a: The nodal pattern for H transfer

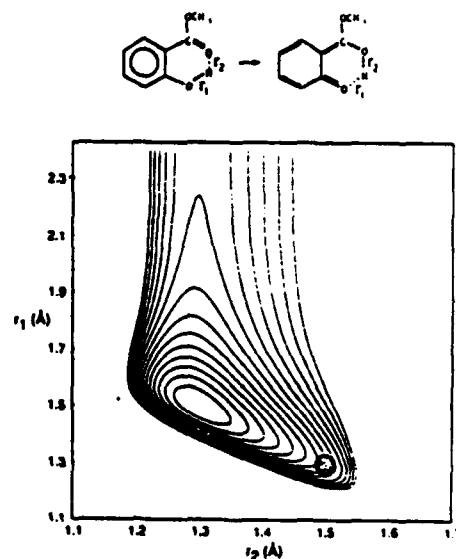


Fig 19b: The wave packet and PES.

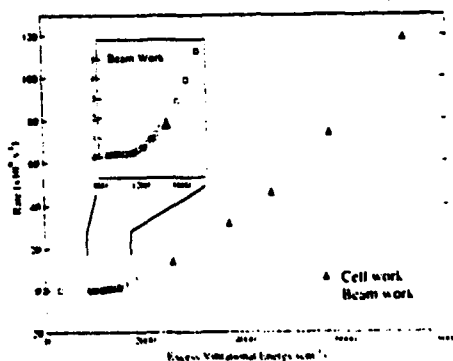


Fig 19 c: The dependence of the rate on energy.

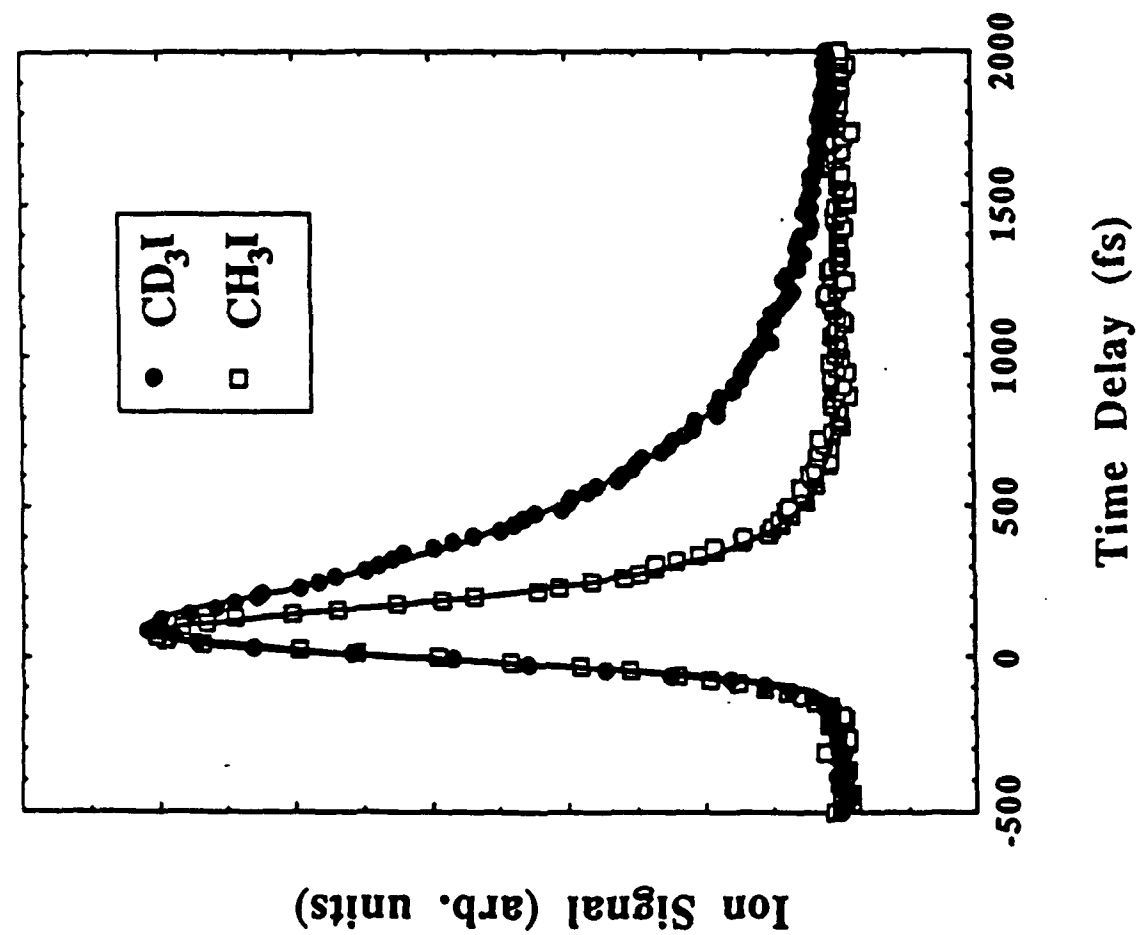


Fig 20: The delay of Rydberg state due to fragmentation : $\text{CH}_3\text{I} \rightarrow \text{CH}_3 + \text{I}$.

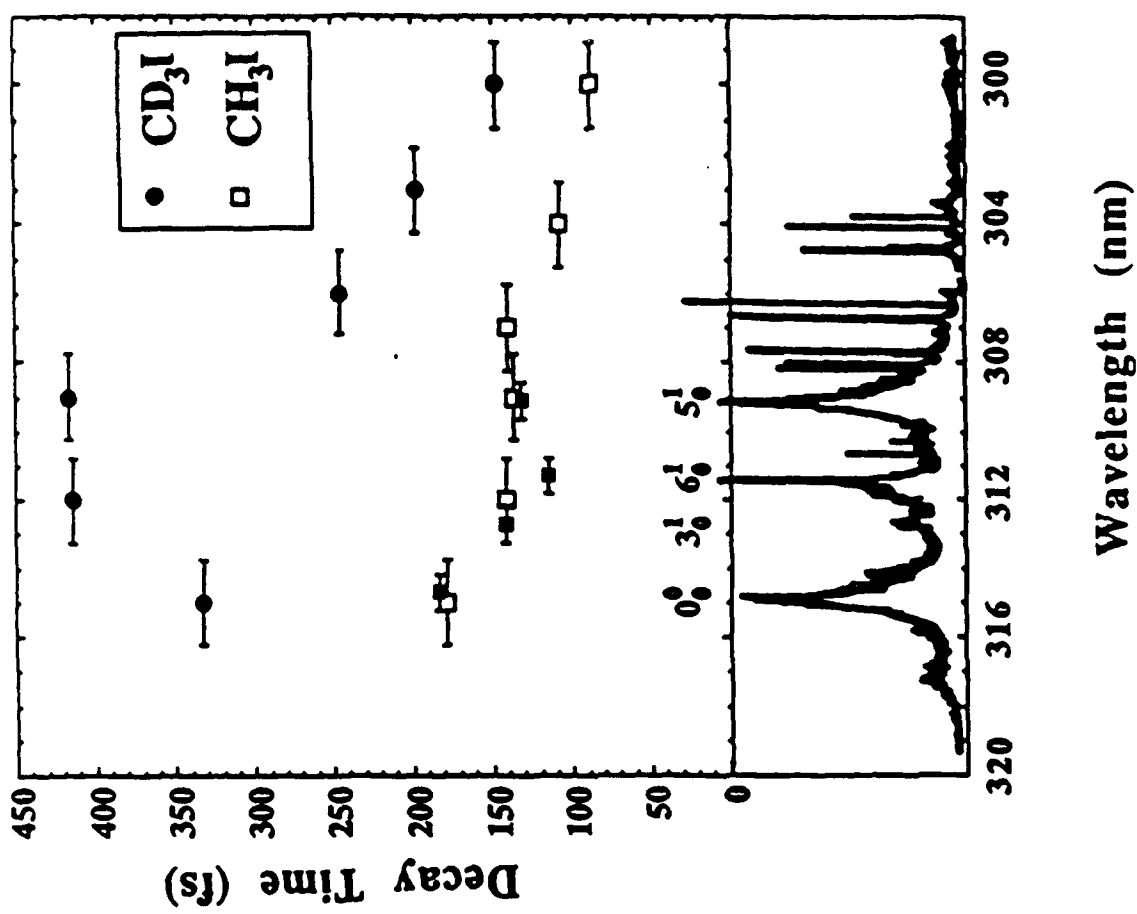


Fig 21: The dependence of the reaction time (dissociation) on energy and character of the mode.

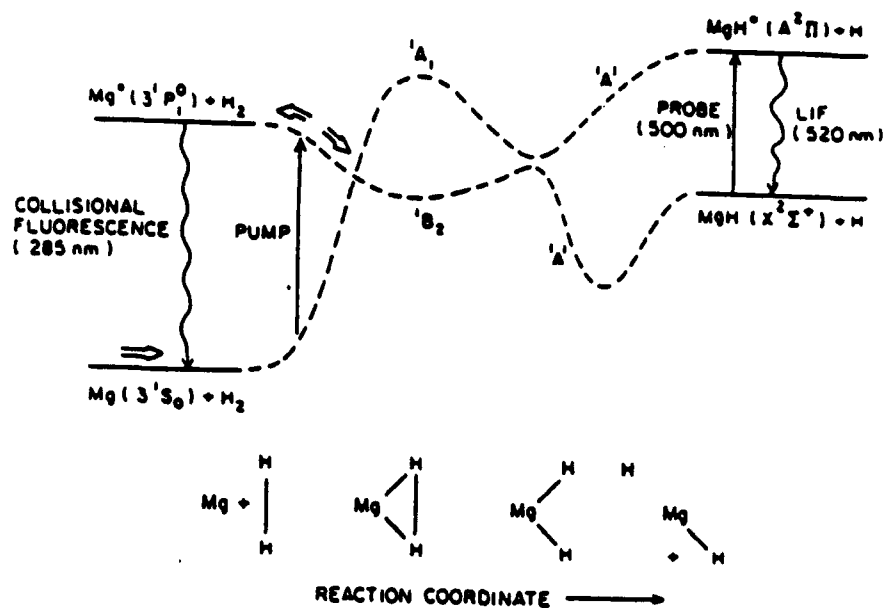


Fig 22: The reactive collision representation for $\text{Mg} + \text{H}_2$, see text.

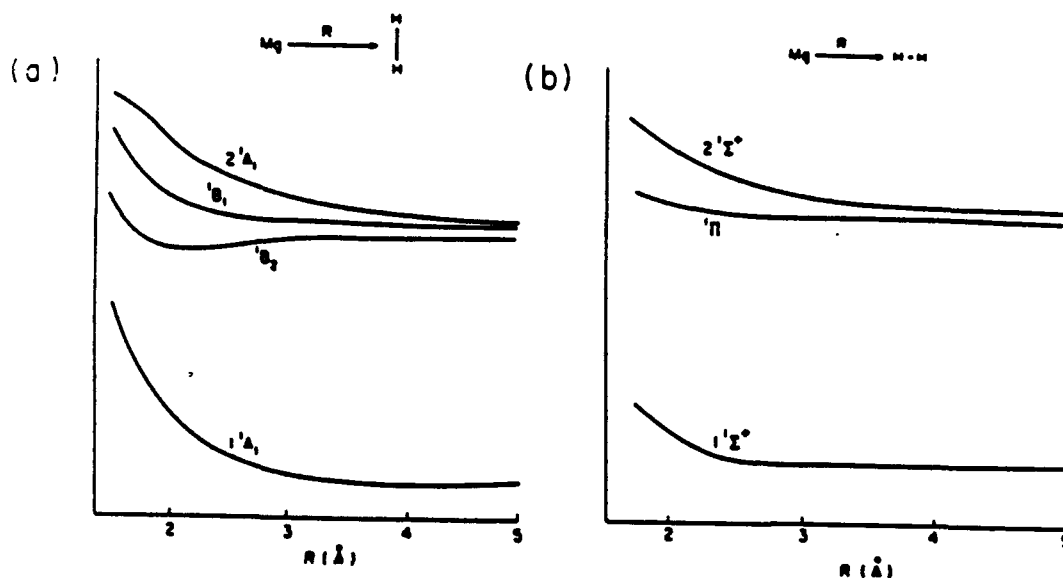


Fig 23: Theoretical potential curves for C_{2v} and $C_{\infty v}$ geometries, text.

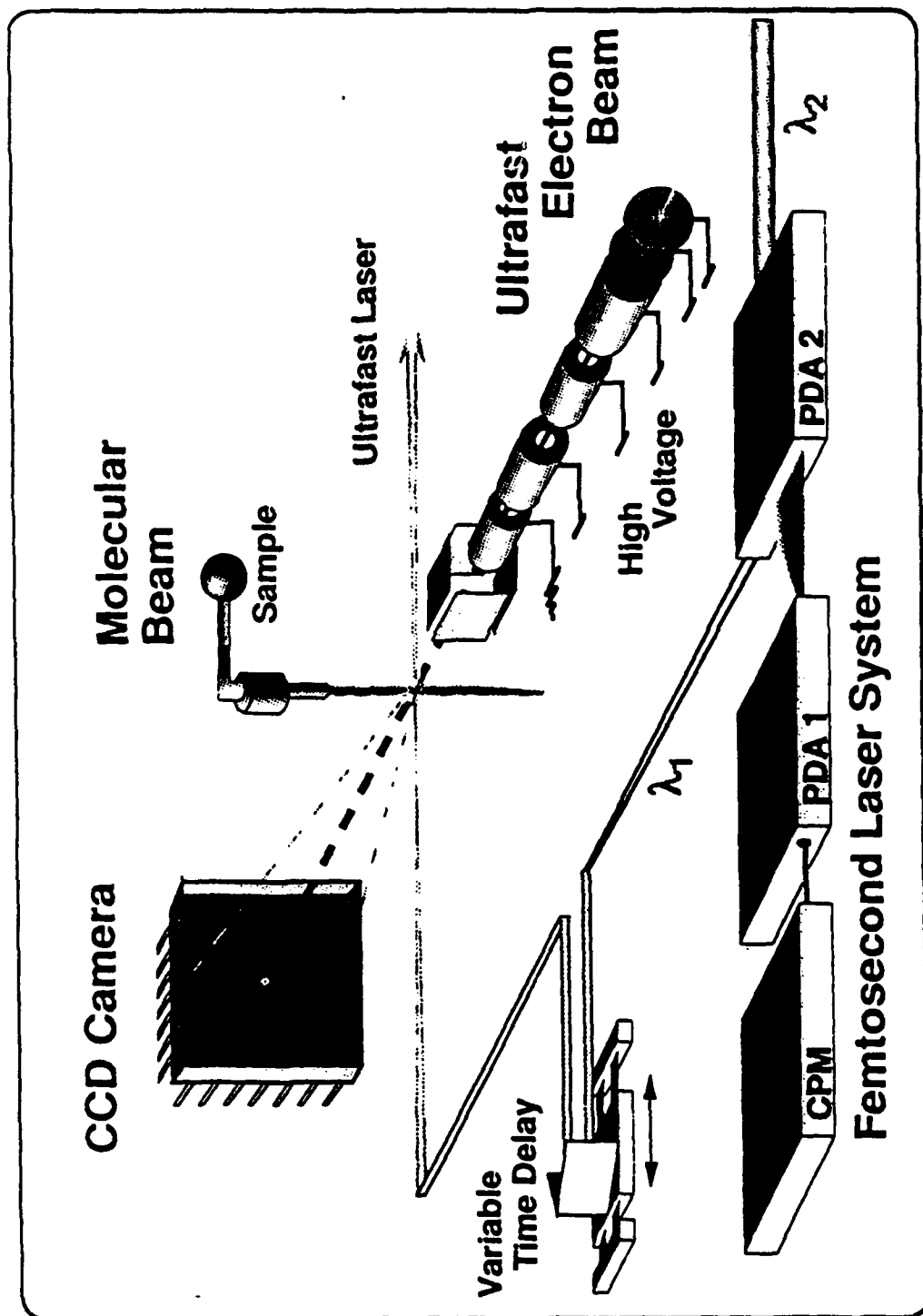


Fig 24: The experimental setup for the electron diffraction apparatus



Fig 25: Diffraction rings as seen by our CCD.

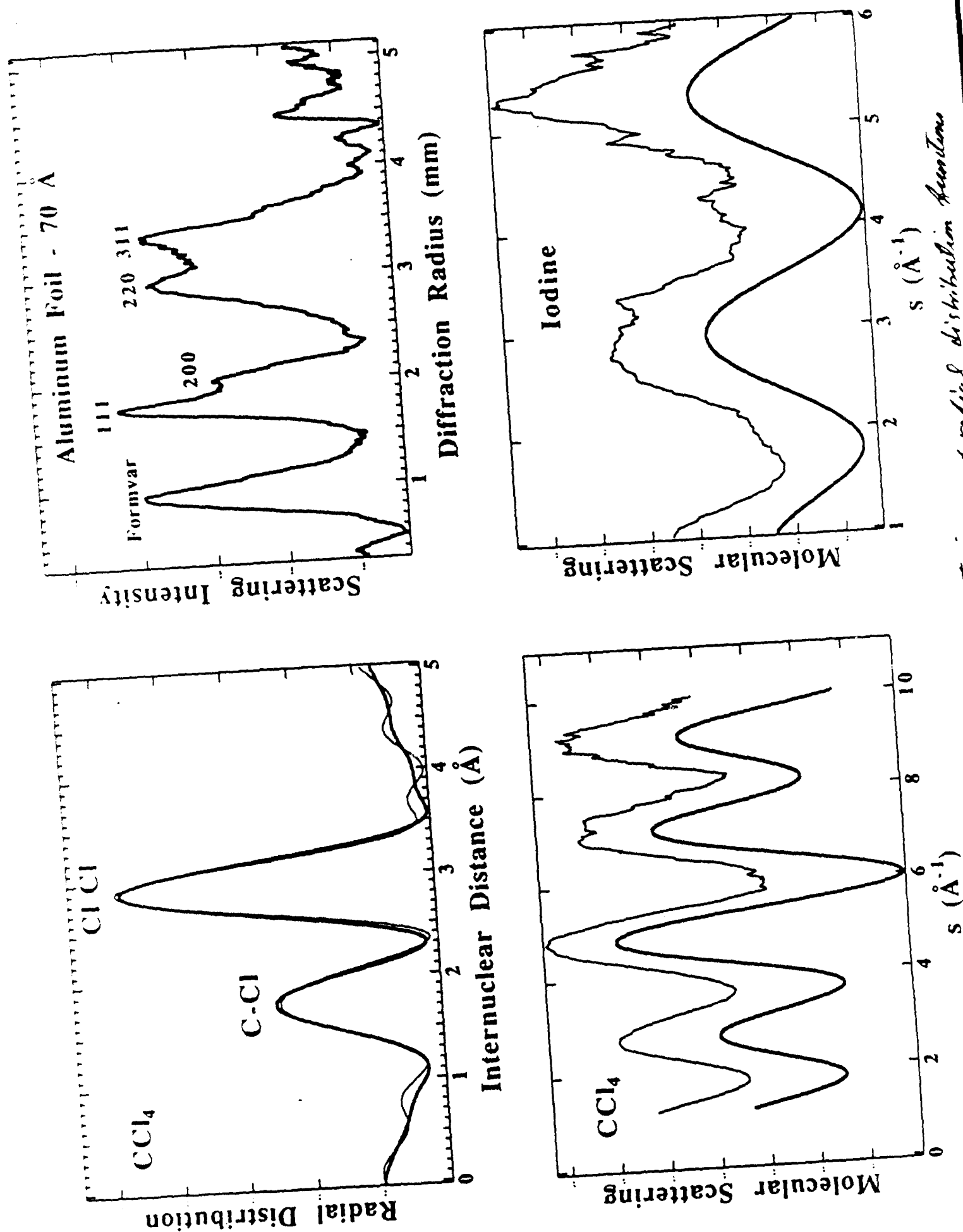


Fig 26: Molecular scattering and radial distribution functions

GRADUATE STUDENTS
AND POST-DOCTORAL RESEARCH FELLOWS

Luis Bañares (postdoc on a Fulbright fellowship from Spain - currently at Complutense de Madrid University)
Spencer Baskin (graduate student - currently at University of Texas)
Thomas Baumert (postdoc on DFG fellowship from Germany - currently at Caltech)
Robert Bowman (postdoc - currently at University of Kansas)
Peijun Cong (graduate student - currently at UC, San Diego)
Marcos Dantus (postdoc - currently at Caltech)
Martin Gruebele (postdoc - currently at University of Illinois)
Jennifer Herek (graduate student - currently at Caltech)
Maurice Janssen (postdoc - currently at University of Amsterdam)
Lutfur Khundkar (graduate student - currently at Northeastern University)
Scott Kim (postdoc - currently at Caltech)
Qianli Liu (graduate student - currently at Caltech)
Amine Mokhtari (postdoc - currently at Ecole Polytechnique)
Søren Pedersen (graduate student - currently at Caltech)
Earl Potter (graduate student - currently at Caltech)
Gareth Roberts (postdoc on SERC fellowship from Cambridge - currently at University of Cambridge)
Todd Rose (postdoc - currently at Aerospace Corporation)
Mark Rosker (postdoc - currently at Rockwell)
Dean Willberg (graduate student - currently at Caltech)
Chuck Williamson (graduate student - currently at Caltech)

BIOGRAPHICAL SKETCH

Ahmed H. Zewail

**Linus Pauling Professor of Chemical Physics
California Institute of Technology, Mail Code 127/72
Pasadena, California 91125 USA**

Personal: Born February 26, 1946, married to Dr. Dema Zewail and have two daughters, Maha (Caltech undergraduate) and Amani (Chandler High School). The current family residence is at 566 Winston Avenue, San Marino, California 91108.

Education: B.S., with honor, Alexandria University (1967); M.S., Alexandria University (1969); Ph.D., University of Pennsylvania (1974); and M.A., h.c., Oxford University.

Professional Experience: Pre-doctoral research fellow, University of Pennsylvania (1969-74); IBM research fellow, University of California, Berkeley (1974-76); Assistant Professor (1976-78), Associate Professor (1978-82), Professor (1982-89), and Linus Pauling Professor of Chemical Physics (1990-present) at Caltech.

Scientific Interests and Contributions: Ultrafast lasers and electron diffraction; femtosecond studies of molecular structure and dynamics in the gas phase, in clusters, and at the interface to solutions. In the past, other areas included: Non-linear laser spectroscopy, dephasing phenomena, and multiple-pulse phase-coherent methods; dynamics of high-energy vibrational local modes; the luminescent solar concentrator (LSC); energy transfer and diffusion in liquids; torsional dynamics of DNA and RNA; disorder and coherence in the solid state; and optical and magnetic properties of excitons. The articles authored and co-authored are at present 250. Editor of five books, the current North American Editor of Chemical Physics Letters, and an editor of the International Series of Monographs on Chemistry (Oxford). Chaired/served (and still) many advisory boards and chaired/co-chaired eight international conferences. Speaker and writer for public science education.

Awards and Honors: Distinction with first-class honor (1967); IBM fellow (1975); Alfred P. Sloan fellow (1978-82); outstanding younger chemist (1981); Fellow of American Physical Society (1982); John Simon

Guggenheim fellow (1987); National Academy of Sciences (1989); Third World Academy of Sciences (1989); The Egyptian-American Person of the Year (1990); Sigma Xi Society (1992).

Camille and Henry Dreyfus Teacher-Scholar Award (1979-85); Alexander von Humboldt Award (1983); National Science Foundation Award (1984, 1988); Buck-Whitney Medal of American Chemical Society (1985); King Faisal International Prize (1989); Harrison-Howe Award of the American Chemical Society (1989); Hoechst Prize (1990); NASA Award (1991); First AAM Achievement Award (1991); Nobel Laureate Signature Award of the American Chemical Society (1992); Carl Zeiss International Award (1992); Medal and Shield of Honor (1992); E. K. Plyler Prize of the American Physical Society (1993); Wolf Prize (1993).

Honorary Lectures include: W. Albert Noyes, Jr. Memorial Lecture (1987); Jean Day Lecture (1988); Francis E. Blacet Lecture (1988); Eyring Lecture (1989); Harry Emmett Gunning Lecture (1989); E. C. Watson Lecture (1990); Max T. Rogers Lecture (1990); Reilly Lecture (1990); Flygare Memorial Lecture (1990); Royal Institution Faraday Public Discourse (1991); Oxford Hinshelwood Lectures (1991); Jacob Bigeleisen Lecture (1991); Richard B. Bernstein Memorial Lecture (1991); Peter A. Leermakers Lecture (1991); University of Birmingham Faculty Lecture (1991); Baxter Lecture (1992); Arthur D. Little Lecture (1992); Zeiss Public Lecture (1992); Mary E. Kapp Lecture (1992); William Pyle Philips Lecture (1992); E. U. Condon Lecture (1992); Royal Society Science and Medicine Lecture (1992); Other plenary and keynote lectures (30), and conference, symposium, and university invited lectures (250) are listed separately.

Honorary Visiting Professor at: University of Amsterdam (1979), University of Bordeaux (1981), Ecole Normale Supérieure (1983), Technical University of Munich (1983), University of California at Los Angeles (1988), American University in Cairo (1988), Johann Wolfgang Goethe-Universität (1990), Oxford University (1991), Texas A&M University (1992), and University of Iowa (1992).